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St Petersburg, Russia, June 18–22, 2001

Fine structure of excitons and e–h pairs in GaAs/AlAs superlattices at the X-Γ crossover

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Abstract. Optically detected magnetic resonance (ODMR) and level anticrossing spectroscopy was applied to study g-factors and exchange splitting of localized excitonsand e–h pairs at the $X_z-\Gamma$ crossover of the conduction band states in a GaAs/AlAs superlattice with a composition gradient. In the transition region we clearly observed disappearance of type II excitons and appearance of type I excitons. In addition, "intermediate" type-II-like and type-I-like excitons were found by ODMR and LAC. Besides ODMR of excitons with a definite fine structure splitting ODMR ascribed to separately localized electrons and holes with a distribution of exchange splittings was detected.

Introduction

In GaAs/AlAs superlattices (SL) both type II and type I band alignment can be obtained. When a GaAs quantum well (QW) thickness is greater than 36 Å, the lowest-energy subbands of the conduction and valence bands are Γ states in the QW (type-I SL). Below a GaAs thickness of Å and provided that the AlAs thickness is not too small the electron and hole wavefunctions are derived from X states in AlAs and Γ states in GaAs and the SL is of type II. The quantum confinement makes the X valley in AlAs split into two states, i.e., X_z along the growth direction [001] and X_{xy} perpendicular to it. Owing to the competition between confinement and strain effects, the lowest electron state is X_z for AlAs thickness below 70 Å or X_{xy} otherwise. Optical transitions are direct in type I systems. In type II SL the optical transitions are indirect and therefore forbidden. For recombination that creates a heavy hole (HH) with Xz electrons the transitions become weakly allowed due to the $\Gamma-X$ coupling (pseudodirect type II SL).

Type II GaAs/AlAs SL's were widely studied by optically detected magnetic resonance (ODMR) which made it possible to measure directly electron and hole g-factors and exciton exchange splitting [1, 2], to determine the order of the conduction band valleys X_z and X_{xy} from the anisotropy of the electron g-factor [3], and also to use these results for a local diagnostics of SL's [4]. Early ODMR studies of the type II-type I transition [5, 6] were mainly focused on the extreme cases of type II and type I recombination. In these works level-anticrossing (LAC) spectroscopy of type I systems was developed [5–7]. In the present communication we apply ODMR and LAC for a study of the $X-\Gamma$ crossover in GaAs/AlAs SL with a smooth transition from pseudodirect type II to type I SL.

1. Results and discussion

Several MBE-grown GaAs/AlAs SL's were studied but the most important results were obtained on a SL (E913) grown with the spatial gradient of the GaAs/AlAs composition in the SL plane along the sample length: from 20.8 Å/12.2 Å to 22.8 Å/11.2 Å. A smooth transition from a pseudodirect type-II SL ($x = 0$) to type-I SL ($x = 23$ mm) has been evidenced in this sample by the time-resolved luminescence measurements [8] and by

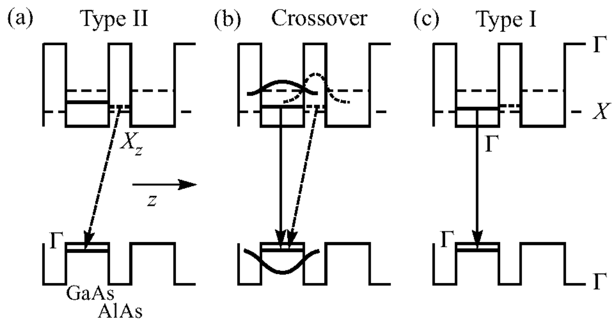

Fig. 1. The valence and conduction band structure together with the recombination transitions for the type II (a), type I (c) and crossover (b) regions of the investigated GaAs/AlAs superlattice with a composition gradient. Narrow lines show energy of Γ(solid lines) and X (dashed lines) states in the bulk materials, thick lines correspond to the superlattice states. In (b), the calculated probability densities of the wavefunctions of electrons and holes in each quantum state are also shown. The axis z is the growth direction.

ODMR and LAC spectroscopy [5–7]. A simplified diagram of the valence and conduction band structure calculated within the envelope wave function approximation using the ECA4 program [9] is shown in Fig. 1 together with the recombination transitions. The energy difference between the X_z and Γ states decreases and reverse its sign, i.e. a $X-\Gamma$ crossover occurs with the change of the GaAs/AlAs composition along the sample. Figure 2 shows the experimental dependences of circularly polarised luminescence on the magnetic field measured at different positions of the excitation spot on the sample with 35 GHz microwaves applied. Luminescence was excited far above the bandgap and detected within the zero-

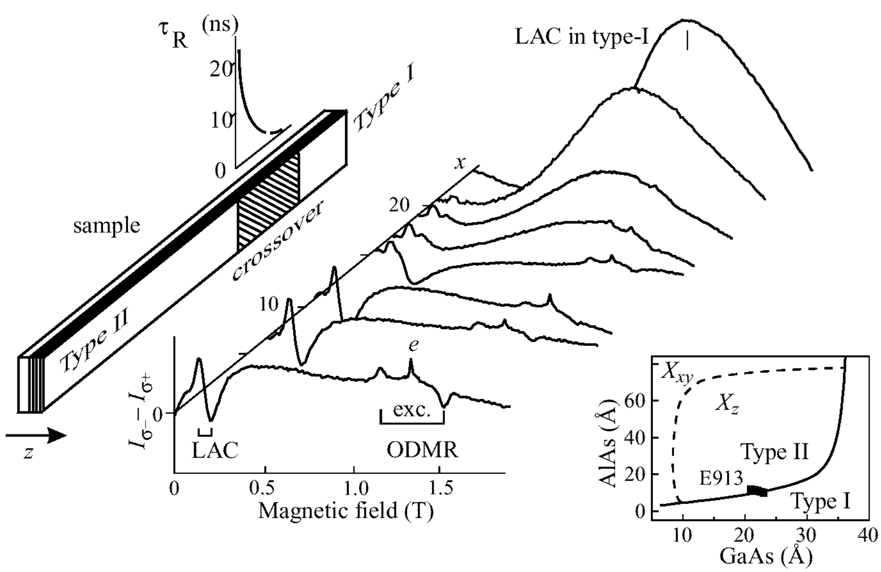

Fig. 2. 35 GHz ODMR and LAC measured as variations of the circular polarization of luminescence at different positions of the excitation spot on the GaAs/AlAs SL (sample E913) with a smooth transition from type II to type I. Inset shows the position of the investigated sample in the map displaying the boundary between type II and type I SL. $T = 1.6$ K. $B \parallel [001]$.

phonon luminescence line. The location of the sample E913 in the map displaying the boundary between type II and type I SL is shown in the insert.

In Fig. 2 resonance signals at 1–1.5 T are due to the microwave-induced EPR transitions between the exciton levels and those of the electrons in e–h pairs. In many cases ODMR of holes was also observed with smaller intensity because for holes the EPR transitions are forbidden and relaxation is much faster. One can see that ODMR disappears in the crossover region ($x = 15-20$ mm) where τ_R decreases from the μs range down to 0.3 ns. In addition to the ODMR, resonance signals produced by the state mixing at anticrossing of optically allowed and forbidden exciton levels are seen in Fig. 2. LAC's do not depend on microwaves and can be observed even for very short lifetimes.

The experimental spectra in a wider field range are shown in Fig. 3. At the type II side of the sample one kind of excitons is observed both by ODMR and LAC. For type I side only LAC can be detected for excitons with about an order of magnitude higher exchange splitting Δ [5–7]. Since the $X-\Gamma$ mixing is proportional to $1/(E_X - E_\Gamma)^2$ it strongly increases in the transition region and new type-II-like and type-I-like excitons appear with the parameters different from the type II and type I excitons ((1) and (2) in Fig. 3).

For type-II-like excitons ODMR is not is detected because of shorter τ_R and the lines of LAC are broadened.

The isotropic exchange splitting $\Delta = 21\,\mu$eV, i.e. larger than $\Delta = 13.3\,\mu$eV for type II excitons in this region, $g_{h\|} = 2.4$, $g_e = 1.8$. For type-I-like excitons the parameters $\Delta = 140\,\mu$eV, $g_{h\|} = 2.4$, $g_e = 1.05$ are close to $\Delta = 170\,\mu$eV, $g_{h\|} = 2.4$, $g_e = 0.9$ for

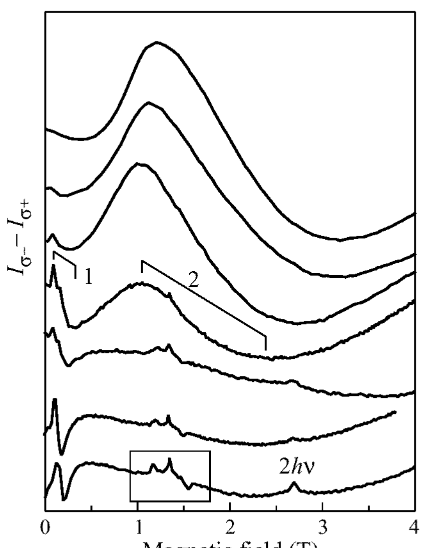

Fig. 3. Disappearance of the characteristic ODMR and LAC of HH–X_z type II excitons and appearance of LAC of HH–Γ type I excitons at the $X-\Gamma$ crossover. (1) and (2) mark the positions of the type-II-like and type-I-like excitons. The spectra were taken at $x = 0$, 8, 15, 17, 19, 21, and 23 mm in SL E913 (from bottom to top).

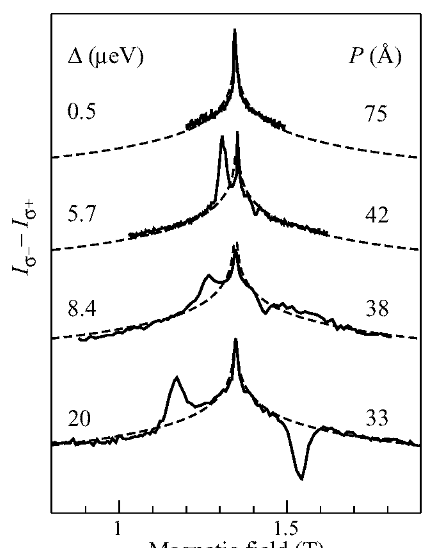

Fig. 4. Coexistence of ODMR originating from type II excitons with a definite exchange splitting D and ODMR of e–h pairs with a distribution of interpair separations in different type II GaAs/AlAs SL's with a period P. The dashed line is calculated for a random distribution of e and h localized in the SL plane. $\nu = 35$ GHz. $B \parallel [001]$.

type I excitons at $x = 23$ mm. At the crossover recombination is determined by an interplay of the recombination rates and probability for the electron to be at the X_z or Γ level. The HH–Γ recombination is faster, therefore coexistence of type-II-like and type-I-like excitons is possible only for X_z below Γ.

Besides the exciton ODMR (" exc." in Fig. 2), broad ODMR signals are seen in Fig. 2 and 3 with $g = 1.88$ corresponding the X_z electrons in AlAs. This ODMR in E913 is shown in the bottom of Fig. 4 in which ODMR recorded in other SL's with different periods P is represented. It is seen that the splitting between the exciton ODMR lines strongly depends on the SL period whereas the shape of the broad ODMR signal does not vary and is period independent in the first approximation. For some SL's we observed similar ODMR of holes. For excitons the splitting of the ODMR lines is proportional to Δ which has a definite value for each SL [3, 7]. Observation of the same shape of the broad electron signal by multiquantum ODMR (the effective frequencies 35, 70, 105 GHz) proves that the broadening is determined by a spreading of the zero-field (i.e. exchange) splittings rather than the g-factors. This implies an existence of e–h recombination of the electrons in AlAs and holes in GaAs which may be localized by the interface roughness potential. Dashed line in Fig. 4 shows the result of calculations for a random distribution of thermalised e–h pairs in the SL plane. For the excitons which are localised as a whole the e–h separation is mainly determined by the SL period. In the $X_z-\Gamma$ transition region ODMR of electrons is seen even after the disappearance of the type-II excitons probably due to longer e–h recombination times.

Acknowledgements

We are indebted to P. Lavallard, C. Gourdon, and R. Planel for supplying some samples and fruitful discussions. This work was supported in part by the Russian Foundation for Basic Research (grant 00-02-16950) and the PSSN program (grant 99-3012).

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